

International Journal of Modern Physics B
 © World Scientific Publishing Company

Exact and approximate relations for the spin-dependence of the exchange energy in high magnetic fields

J. M. Morbec* and K. Capelle

*Departamento de Física e Informática
 Instituto de Física de São Carlos
 Universidade de São Paulo
 Caixa Postal 369, 13560-970 São Carlos, SP, Brazil*

The exchange energy of an arbitrary collinear-spin many-body system in an external magnetic field is a functional of the spin-resolved charge and current densities, $E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow]$. Within the framework of density-functional theory (DFT), we show that the dependence of this functional on the four densities can be fully reconstructed from either of two extreme limits: a fully polarized system or a completely unpolarized system. Reconstruction from the limit of an unpolarized system yields a generalization of the Oliver-Perdew spin scaling relations from spin-DFT to current-DFT. Reconstruction from the limit of a fully polarized system is used to derive the high-field form of the local-spin-density approximation to current-DFT and to magnetic-field DFT.

Keywords: density-functional theory, exchange energy, orbital magnetism

1. Introduction

The exchange energy is a fundamental quantity of many-body physics. While it can be calculated analytically and exactly for simple quantum fluids, such as the three-dimensional interacting electron gas in the absence of magnetic fields, its evaluation for realistic, spatially inhomogeneous, many-electron systems is possible only numerically, *e.g.* by means of density-functional theory (DFT) within the local-spin-density approximation (LSDA).^{1,2} In collinear spin-DFT (SDFT), the exact exchange energy is a functional of the spin-resolved charge density $E_x[n_\uparrow, n_\downarrow]$. In strong external or internal magnetic fields, orbital magnetism must be accounted for on the same footing as spin magnetism, and SDFT is replaced by current-DFT (CDFT)^{3,4} or magnetic-field DFT (BDFT).^{5,6}

In the present paper we discuss exact and approximate expressions for the spin-dependence of the exchange energy E_x . We show that, for arbitrary collinear-spin many-body systems, the dependence of E_x on spin-resolved charge and current densities can be fully reconstructed from either of two extreme limits: a fully polarized system or a completely unpolarized system. Reconstruction from the limit of an unpolarized system yields a generalization of the Oliver-Perdew spin scaling relations⁷

*Communicating author. Email: jmorbec@yahoo.com.br

from SDFT to CDFT. The alternative possibility of reconstruction from the limit of a fully polarized system results in explicit expressions for the high-field exchange energy, which can be used as input for the LSDA of CDFT or of BDFT.

2. Exact properties: spin scaling

Since exchange acts only between like-spin electrons, the exchange energy of an arbitrary many-electron system (homogeneous or not, in a magnetic field or not) can be written as the sum of two contributions, one arising entirely from the spin up subsystem, the other from the spin down subsystem

$$E_x = \tilde{E}_x^\uparrow + \tilde{E}_x^\downarrow. \quad (1)$$

This decomposition is valid as long as there is a preferred direction relative to which one can define spin up and spin down, *i.e.* for collinear magnetism. It only ceases to be valid in noncollinear situations, where the quantization axis changes as a function of space. The exchange energy then acquires a spin off-diagonal contribution which, in principle, can be treated on the same footing as the diagonal term.⁸ Here, we are only concerned with collinear situations, occurring in unidirectional magnetic fields. Under these circumstances, Eq. (1) always holds.

According to the general theorems of CDFT,^{3,4} the exchange energy of an arbitrary (collinear) many-body system is a functional of the spin-resolved charge and current densities $n_\uparrow(\mathbf{r})$, $n_\downarrow(\mathbf{r})$, $\mathbf{j}_\uparrow(\mathbf{r})$ and $\mathbf{j}_\downarrow(\mathbf{r})$, where $\mathbf{j}(\mathbf{r}) = \mathbf{j}_\uparrow(\mathbf{r}) + \mathbf{j}_\downarrow(\mathbf{r})$ is the paramagnetic current density, related to the gauge invariant physical current density by

$$\mathbf{j}_{\text{phys}}(\mathbf{r}) = \mathbf{j}(\mathbf{r}) + \frac{e}{mc} n(\mathbf{r}) \mathbf{A}(\mathbf{r}). \quad (2)$$

By symmetry, $E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow] = E_x[n_\downarrow, n_\uparrow, \mathbf{j}_\downarrow, \mathbf{j}_\uparrow]$. The decomposition (1) then implies

$$E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow] = \tilde{E}_x[n_\uparrow, \mathbf{j}_\uparrow] + \tilde{E}_x[n_\downarrow, \mathbf{j}_\downarrow], \quad (3)$$

where the spin up and spin down contribution are given by the same functional, although their values for specific densities, \tilde{E}_x^\uparrow and \tilde{E}_x^\downarrow , are, generally, different.

We now define two related functionals, the exchange energy of a spin unpolarized system⁹

$$E_x^{NP}[n, \mathbf{j}] := E_x \left[\frac{n}{2}, \frac{n}{2}, \frac{\mathbf{j}}{2}, \frac{\mathbf{j}}{2} \right] \quad (4)$$

and that of a fully polarized system

$$E_x^{FP}[n, \mathbf{j}] := E_x[n, 0, \mathbf{j}, 0]. \quad (5)$$

By evaluating Eq. (3) at the densities of an unpolarized system, we obtain

$$\tilde{E}_x[n, \mathbf{j}] = \frac{1}{2} E_x^{NP}[2n, 2\mathbf{j}], \quad (6)$$

from which it follows that we can write E_x as

$$E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow] = \frac{1}{2}E_x^{NP}[2n_\uparrow, 2\mathbf{j}_\uparrow] + \frac{1}{2}E_x^{NP}[2n_\downarrow, 2\mathbf{j}_\downarrow], \quad (7)$$

which holds for arbitrary polarizations. By evaluating this at the densities of a fully polarized system, we find $\tilde{E}_x[n, \mathbf{j}] = E_x^{FP}[n, \mathbf{j}]$, which implies that the general exchange energy can also be written as

$$E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow] = E_x^{FP}[n_\uparrow, \mathbf{j}_\uparrow] + E_x^{FP}[n_\downarrow, \mathbf{j}_\downarrow]. \quad (8)$$

The two limiting cases of the general functional $E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow]$ are thus related by

$$E_x^{FP}[n, \mathbf{j}] = \frac{1}{2}E_x^{NP}[2n, 2\mathbf{j}]. \quad (9)$$

Equations (7) and (8) show that the exchange energy has the remarkable property that its functional form at arbitrary polarization is completely determined by either that at zero polarization or that at full polarization, without requiring interpolation between both limits or additional calculations inbetween them.

If its current dependence is ignored, Eq. (7) reduces to the spin-scaling relations of Oliver and Perdew,⁷ which are frequently used in spin-density-functional theory (SDFT) to connect spin-density functionals for arbitrarily polarized systems to density functionals obtained from many-body calculations for an unpolarized system.^{10,11,12} Equation (7) is the generalization of this spin scaling from SDFT to CDFT, and may be employed in the same way.

Equations reconstructing the general functional from the fully polarized limit, such as Eq. (8), are less used in DFT, where one usually constructs a spin-dependent functional by starting from the unpolarized situation. However, they are useful in the special case of the high-field limit, where analytical results for the fully polarized exchange energy of the homogeneous interacting electron gas are available for the first ($L = 0$)¹³ and second ($L = 1$)¹⁴ Landau level.

3. Approximate expressions: high-field exchange energy of the homogeneous electron gas

In the homogeneous electron gas in a constant unidirectional magnetic field B , the charge density and the physical current density are spatially constant. For sufficiently high magnetic fields only the lowest Landau level contributes, and the electrons are fully spin polarized. The per-volume exchange energy $e_x^{FP}(n, B) = E_x^{FP}(n, B)/V$ in this situation has been evaluated analytically by Danz and Glasser.¹³ In the limit in which all electrons occupy the bottom of the lowest Landau level, $L = 0$, they find,

$$e_x^{FP}(n, B) = 2\pi e^2 \lambda^2 n^2 [\ln(\lambda^3 n) + 2.11779], \quad (10)$$

where $\lambda = (\hbar c/eB)^{1/2}$ is the magnetic length.

Expressions applicable to still higher fields, as well as generalization to weaker fields in which the restriction to the bottom of the lowest Landau level is removed

(while maintaining that to the lowest Landau level, $L = 0$, itself) can be found in the original reference.¹³ In a recent paper¹⁴ we reevaluated the exchange energy in these limits, corrected a few minor problems in the original equations,^{13,15} and discussed the range of magnetic fields and densities where each of them applies.

For still weaker magnetic fields, two complications can take place: orbital magnetism now acquires a contribution of higher Landau levels, and spin magnetism is reduced, as the system is not necessarily fully polarized. We dealt with the former complication by deriving an explicit expression for $L = 1$.¹⁴ Here, we consider partial and vanishing spin polarization, which may occur for lower fields, in particular when the electron g factor is smaller than 2.

In a first step, we note that the many-body calculations^{13,14,15} yield $e_x^{FP}(n, \mathbf{B})$ as function of the charge density n and the magnetic field B , *i.e.* the variables used in BDFT, but not of the spin and current densities used in CDFT. The transition from one set of variables to the other is affected by recalling that in the homogeneous electron gas in a constant magnetic field $\mathbf{j}_{\text{phys}}(\mathbf{r}) \equiv 0$, so that, from Eq. (2), $\mathbf{B}(n, \mathbf{j}) = -\frac{mc}{e}\nabla \times (\mathbf{j}/n)$. From Eq. (8) we thus have¹⁶

$$e_x(n_{\uparrow}, n_{\downarrow}, \mathbf{j}_{\uparrow}, \mathbf{j}_{\downarrow}) = e_x^{FP}\left(n_{\uparrow}, \mathbf{B} = -\frac{mc}{e}\nabla \times \left(\frac{\mathbf{j}_{\uparrow}}{n_{\uparrow}}\right)\right) + e_x^{FP}\left(n_{\downarrow}, \mathbf{B} = -\frac{mc}{e}\nabla \times \left(\frac{\mathbf{j}_{\downarrow}}{n_{\downarrow}}\right)\right). \quad (11)$$

Equation (11) provides the exchange energy for arbitrarily polarized systems, as a function of the spin-resolved charge and current densities. As an example, we use it to generate from Eq. (10) the exchange energy of an unpolarized system. This can be done either by using (11) to generate the result for an arbitrarily polarized system and subsequently substitute the densities of an unpolarized system, or, equivalently, directly from the special case of Eq. (9). The result is

$$e_x^{NP}(n, \mathbf{B}) = \pi e^2 \lambda^2 n^2 [\ln(\lambda^3 n) + 1.42464]. \quad (12)$$

All other relations, valid for different magnetic field ranges,^{13,14} can be treated in the same way. We have chosen to exemplify the procedure for Eq. (10), because this allows an independent numerical test of our previous assertion (which was based on explicit calculation¹⁴) that the Danz-Glasser expression (10) is correct, and not the alternative result¹⁵

$$e_x^{FP,alt}(n, \mathbf{B}) = 2\pi e^2 \lambda^2 n^2 [\ln(\lambda^3 n) + 2.32918], \quad (13)$$

which, when scaled to the unpolarized case, becomes

$$e_x^{NP,alt}(n, \mathbf{B}) = \pi e^2 \lambda^2 n^2 [\ln(\lambda^3 n) + 1.63603]. \quad (14)$$

In the unpolarized case, Takada and Goto¹⁷ numerically calculated e_x , so that we can use their data to check the validity of Eqs. (12) and (14), and thus also of (10) and (13). We find that (12) is in perfect agreement with numerical data extracted from figure 2(b) of Takada and Goto,¹⁷ whereas (14) deviates from the numerical data by $\sim 10\%$ in the metallic density range.

4. Conclusions

Two types of spin-scaling relations have been demonstrated for the current-dependent exchange energy, one employing the fully polarized limit and the other the unpolarized limit. Simple connections, valid for arbitrary many-body systems, exist between these limits and the general functional $E_x[n_\uparrow, n_\downarrow, \mathbf{j}_\uparrow, \mathbf{j}_\downarrow]$. In the special case of the homogeneous electron gas, such connections, applied to many-body calculations in the fully polarized limit,^{13,14,15} yield explicit formulas that allow to use available numerical data for unpolarized systems¹⁷ to arbitrate between conflicting expressions at full polarization. Results for arbitrary polarization can be used as input for the local-spin-density approximation to current-density-functional theory.

Acknowledgements

This work received financial support from FAPESP and CNPq.

References

1. U. von Barth and L. Hedin, *J. Phys. C* **5**, 1629 (1972).
2. O. Gunnarsson and B. I. Lundqvist, *Phys. Rev. B* **13**, 4274 (1976).
3. G. Vignale and M. Rasolt, *Phys. Rev. Lett.* **59**, 2360 (1987).
4. G. Vignale and M. Rasolt, *Phys. Rev. B* **37**, 10685 (1988).
5. C. J. Grayce and R. A. Harris, *Phys. Rev. A* **50**, 3089 (1994).
6. F. R. Salsbury and R. A. Harris, *J. Chem. Phys.* **107**, 7350 (1997).
7. G. L. Oliver and J. P. Perdew, *Phys. Rev. A* **20**, 397 (1979).
8. K. Capelle and L. N. Oliveira, *Phys. Rev. B* **61**, 15228 (2000).
9. NP standing for “not polarized” — the more natural choice of UP (unpolarized) may lead to confusion with the spin up contribution.
10. J. P. Perdew, A. Ruzsinszky, J. Tao, V. N. Staroverov, G. E. Scuseria, and G. I. Csonka, *J. Chem. Phys.* **123**, 062201 (2005).
11. J. P. Perdew, K. Burke and M. Ernzerhof, *Phys. Rev. Lett.* **77**, 3865 (1996).
12. K. Capelle and G. Vignale, *Phys. Rev. Lett.* **86**, 5546 (2001).
13. R. W. Danz and M. L. Glasser, *Phys. Rev. B* **4**, 94 (1971).
14. J. M. Morbec and K. Capelle, *Phys. Rev. B* **78**, 085107 (2008).
15. B. Banerjee, D. H. Constantinescu and P. Rehak, *Phys. Rev. D* **10**, 2384 (1974).
16. Note that we do not distinguish functionals of the current from those of the magnetic field by a new symbol, since they can always be distinguished through their arguments.
17. Y. Takada and H. Goto, *J. Phys. Condens. Matter* **10**, 11315 (1998).